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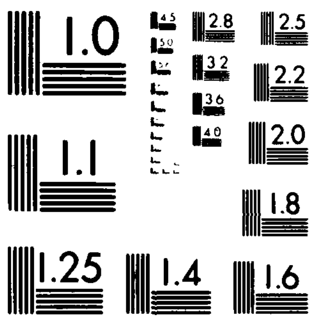

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THEORETICAL STUDIES OF RYDBERG ATOM COLLISIONS

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SRI Project 8467
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Rydberg atom collisions

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Theoretical studies were performed on a variety of topics related to collisions involving Rydberg atoms. Progress was made towards the understanding of ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom *collisions*. A strong dc electric field was also incorporated in calculations of electron capture and ionization cross sections for ion-Rydberg atom collisions.

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ABSTRACT

During the last two years, theoretical studies were performed on a variety of topics related to collisions involving Rydberg atoms. Progress was made towards the understanding of ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom collisions. Cross sections were calculated for electron capture, ionization, and excitation processes and parametrized in terms of atomic parameters. A strong dc electric field was also incorporated in calculations of electron capture and ionization cross sections for ion-Rydberg atom collisions. The results of this latter investigation indicate the design of a far ir photon detector which is based on the field ionization of Rydberg atoms may be limited by the collision processes.

BACKGROUND

At present, it is possible to detect single photons having wavelengths < 1 μm with single quantum detectors. Conventional methods fail at longer wavelengths, because the reduced quantum efficiency leads to signal-to-noise problems. However, because Rydberg atoms have large absorption cross sections for long-wavelength radiation (> 10 μm), these atoms can be used to convert infrared and microwave photons into either visible photons or ions that can be detected by conventional techniques. The basic idea of a Rydberg atom infrared or microwave detector is to make a target of these atoms that is optically thick to the radiation to be detected at wavelength λ_o. The photons with wavelength λ_o will be absorbed in the target of Rydberg atoms in a single n,ℓ quantum state. The atoms that absorb the photons undergo a transition to a state of different parity and energy. This state can then easily be detected by taking advantage of either the different wavelength of the optical radiation, which is subsequently emitted by the atom, or the difference in the field ionization of the two states.

The first question that must be asked is: Is it possible to make an optically thick target of Rydberg atoms for long-wavelength radiation? Recent work[1-3] has demonstrated that the use of Rydberg atoms for detection of infrared and microwave radiation is, in principle, quite feasible. Moreover, it can be orders of magnitude more sensitive than other available detectors, a condition that can lead to a variety of possible applications. It has been shown that it is necessary to produce a 10^8 cm^{-3} density of Rydberg atoms in a parent gas density of $\sim 10^{11}$ cm^{-3} in order to obtain a 1-cm-long optically thick target of Rydberg atoms. This density of Rydberg atoms is now being produced with dye laser technology in several laboratories.

Because it is apparent that the detector will work in principle, it is necessary to determine its limitations. Obvious problems are collisional processes that either destroy the population of the Rydberg atom in a specific (n,ℓ) electronic level, thereby reducing the steady-state population density, or induce the same transition that is used to observe the long-wavelength radiation, thereby producing spurious signals. For either case,

the collision mean free time must be comparable to the radiative lifetime of the Rydberg atom that has been observed[4] to be empirically given by

$$\tau = \frac{n^3}{2.4 \times 10^{+8}} \text{ s} \quad . \tag{1}$$

As an example of the magnitudes of the collisional deactivation cross sections that are necessary to compete with the radiative lifetime, one can use a system in which the Rydberg atom is in the $n = 20$ level. From Eq. (1), one finds that the Rydberg atom has a radiative lifetime of 3.3×10^{-5} s. If a Rydberg atom density of 10^8 cm^{-3} in a ground state density of 10^{11} cm^{-3} is realistically assumed, it is easy to show that collisional deactivation would be comparable to the radiative process if the Rydberg atom-ground state atom deactivation cross section were on the order of 10^{-11} cm^2. Because the geometric cross section for a Rydberg atom is $\pi n^4 a_o^2$ for a Rydberg atom that is in the $n = 20$ level, the deactivation cross section conceivably could be comparable to the geometric cross section, or $\sim 1 \times 10^{-11}$ cm^2. Such a cross section is extremely plausible, especially for alkali atoms, which have large electron scattering lengths and high dipole polarizabilities and can thus easily induce the Rydberg electron to other close-lying electronic levels. It appears, therefore, the collisional processes must be seriously considered in the design of any Rydberg atom long-wavelength photon detector.

During the last two years, we have begun a series of theoretical studies directed towards Rydberg atom collisional processes. This work includes ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom scattering along with collisions in strong electric fields. The results of the work are briefly described in the next section.

RESEARCH PROGRESS

In the last two years, we have focused our attention on several Rydberg atom collision problems. Originally, our theoretical techniques were directed toward ionization collisions involving two Rydberg atoms in the same principal quantum numbers n:

$$A^{***}(n) + A^{**}(n) \rightarrow A^{+} + \ldots \quad . \tag{2}$$

A four-body classical-trajectory Monte Carlo (CTMC) code was written and applied to Reaction (2) for collision velocities, v/v_e, from 10^{-2} to 10^{1} (note: $v_e = 1/n$ a.u. $= 2.2 \times 10^{8}/n$ cm/s). It is of particular interest that the ionization cross sections for (2) were approximately an order of magnitude larger than the geometric value $\pi n^4 a_o^2$ at thermal energies. Also, the CTMC code inherently predicted the importance of the dipole-induced dipole forces at low velocities and showed that the cross section increases as the velocity decreased, as $v^{-2/3}$. The work was published under the title "Ionization Cross Sections by Rydberg-Atom-Rydberg-Atom Collisions" in Phys. Rev. Lett., 43, 126 (1979).

The CTMC code was also applied to collisions of ions with Rydberg atoms in the v/v_e range of 1 to 10. Both electron capture,

$$A^{+q} + B(n) \rightarrow A^{+q}(n') + B^{+} \quad , \tag{3}$$

and ionization,

$$A^{+q} + B(n) \rightarrow A^{+q} + B^{+} + e^{-} \quad , \tag{4}$$

cross sections were calculated. The cross sections were conveniently presented in terms of simple analytical expressions containing the collision velocity, incident ion charge state, and electronic level of the Rydberg atom. The most interesting aspect of the calculations was the determination of the $A^{+q-1}(n')$ product ion distributions after the electron capture

reaction (3). The net result was that the Rydberg atoms's electron after capture tries to preserve its original dimensions and orbital energy. Consequently, the most probable final state can be expressed by

$$n' = nq^{3/4} \quad . \tag{5}$$

The work was published under the title "Ion-Rydberg Atom Collision Cross Sections" in J. Phys. B, 13, 483 (1980).

In the area of ion-Rydberg atom excitation transfer collisions,

$$A^{+q} + B(n) \rightarrow A^{+q} + B(n') \quad , \tag{6}$$

we recently completed benchmark calculations on the $N^{+3} + H^{**}(n)$ system in the range of $n = 9$ to 24. These calculations were motivated by the work of Kim and Meyer[3] who ignored the excitation transfer process in the analysis of their data and incorrectly thought that their observed cross sections were due to the ionization process. A paper that is concerned with the excitation transfer process, "Excitation Transfer in Ion-Rydberg Atom Collisions," has been accepted for publication in The Physical Review.

A major portion of our time during 1980 has been spent in writing a review on theoretical methods and results as applied to Rydberg atom collisions. The review covers "ℓ-changing" collisions between Rydberg atoms and ground state neutral atoms, associative ionization processes between Rydberg and ground state atoms, ion-Rydberg atom collisions, and Rydberg atom-Rydberg atom processes. It will be included as a chapter in a book, entitled Rydberg Atoms, which is to be edited by R. Stebbings and published by Cambridge University Press. Our chapter will be entitled: "Theoretical Approaches to Low Energy Collisions of Rydberg Atoms with Atoms and Ions" and will be co-authored with A. P. Hickman and J. Pascale.

An interesting research topic for which we have completed preliminary calculations is the effect of strong dc electric fields on ion-Rydberg atom ionization and electron capture cross sections. This is a problem that is extremely pertinent to a far infrared photon detector that is based on Rydberg atoms, with the use of field ionization to determine the product state. In fact, because the cross sections are so highly dependent on the magnitude of

the electric field, these cross sections may determine the efficiency of the field ionization in the detector.

The electric field calculations were accomplished by the CTMC method. Model problems on Rydberg atoms in the $n = 10$ and $n = 20$ states were solved, and the calculated cross sections were parameterized in terms of the quantum levels of the Rydberg atom, the electric field strength, and the collision velocity. Interestingly, the electric field caused the cross sections for electron capture to decrease by up to fourfold, while the ionization values increased by up to two orders of magnitude. A paper entitled "Ion Collisions with Rydberg Atoms in Strong Electric Fields," which is co-authored with A. D. Mackellar, has been accepted for publication in Physical Review Letters.

A paper describing work that was partially supported by ONR has been submitted for publication in the Journal of Physics B (author: A. P. Hickman). It describes quantum close-coupling calculations that investigate the importance of Rydberg atom core interactions in "ℓ-mixing" collisions with rare gases.

PUBLISHED WORK

The titles and authors of the papers attributed to this contract over the last two years are listed below. The covering pages to the papers follow.

a. Ionization Cross Sections for Rydberg Atom-Rydberg Atom Collisions, R. E. Olson, Phys. Rev. Lett. 43, 126 (1979).

b. Ion-Rydberg Atom Collision Cross Sections, R. E. Olson, J. Phys. B 13, 483 (1980).

c. Theoretical Approaches to Low Energy Collisions of Rydberg Atoms with Atoms and Ions, A. P. Hickman, R. E. Olson, and J. Pascale, chapter for Rydberg Atoms, to be published by Cambridge University Press and edited by R. F. Stebbings.

d. Excitation Transfer in Ion-Rydberg Atom Collisions, R. E. Olson, Phys. Rev. A (accepted).

e. Ion Collisions with Rydberg Atoms in Strong Electric Fields, R. E. Olson and A. D. Mackellar, Phys. Rev. Lett. (accepted).

f. The Effect of Core Interactions in ℓ-mixing Collisions of Rydberg Atoms with Rare Gases, A. P. Hickman, J. Phys. B (submitted).

ABSTRACTS FROM PUBLICATIONS

ABSTRACTS FROM PUBLICATIONS

a.

VOLUME 43, NUMBER 2 PHYSICAL REVIEW LETTERS 9 JULY 1979

Ionization Cross Sections for Rydberg-Atom–Rydberg-Atom Collisions

R. E. Olson
Molecular Physics Laboratory, SRI International, Menlo Park, California 94025
(Received 23 April 1979)

A classical-trajectory Monte Carlo method has been applied to collisions of two Rydberg atoms. Numerical calculations were made for velocities $v = 0.01v_e$ to $10v_e$, where the Rydberg electron's velocity $v_e(\text{a.u.}) = 1/n$ and n is the principal quantum number of the Rydberg atom. The total ionization cross sections scale as n^4 and show a $v^{-0.65}$ dependence at low v, a slight maximum around v_e, and a rapid decrease at high v. The cross sections are almost an order of magnitude larger than $\pi n^4 a_0^2$ at thermal energies.

b.

J. Phys. B: Atom. Molec. Phys. 13 (1980) 483–492. Printed in Great Britain

Ion–Rydberg atom collision cross sections

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Received 19 June 1979, in final form 29 August 1979

Abstract. Classical-trajectory Monte Carlo calculations have been performed for collisions of ions in charge states $q = +1, +2, +5$ and $+10$ with hydrogenic atoms in principal quantum levels $n = 1, 2, 5, 10$ and 20. The collision velocity range investigated was $1 \leq v/v_e \leq 10$ where v_e is the orbital velocity of the Rydberg electron ($1/n$ in atomic units). Both charge-exchange and impact ionisation cross sections were calculated with impact ionisation found to be the dominant channel for $v/v_e \geq 2$. For $v/v_e \geq 5$, the sum of the charge-exchange (CEX) and impact ionisation (ION) cross sections may be represented by $\sigma_{\text{CEX+ION}}(a_0^2) = 6\pi n^2 q^2/v^2$, where v is in atomic units. Analysis of the electronic levels produced after charge exchange by the ion indicates the capture proceeds into excited levels which tend to preserve the energy and orbital size of the initial Rydberg atom.

c. Chapter for : Rydberg Atoms edited by R. F. Stebbing and published by Cambridge University Press

THEORETICAL APPROACHES TO LOW ENERGY COLLISIONS OF RYDBERG ATOMS WITH ATOMS AND IONS

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CONTENTS

d. Phys. Rev. A (accepted)

Excitation transfer in ion–Rydberg-atom collisions

R. E. Olson*

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(Received 11 September 1980)

Recently, electron-loss cross sections were presented by Kim and Meyer [Phys. Rev. Lett. 44, 1047 (1980)] for 40 keV/amu N^{5+} + $H^{**}(n)$ collisions which scaled as $n^{3/2}$, where n is the principal quantum number of the excited H^0. Such results are in contrast to an n^2 scaling predicted by classical and first Born theoretical methods. Our calculations indicate that a major component of the experimentally observed ion signal was due to Stark ionization by deflector grids of highly excited H^0 produced in excitation-transfer collisions. Inclusion of the excitation process in a theoretical interpretation reveals qualitative agreement between theory and experiment and stresses the importance of excitation transfer in ion–Rydberg-atom collisions.

e. Phys. Rev. Lett. (accepted)

ION COLLISIONS WITH RYDBERG ATOMS IN STRONG ELECTRIC FIELDS

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ABSTRACT

The classical-trajectory Monte Carlo method has been used to investigate collisions of ions and Rydberg atoms in strong dc electric fields. Cross sections are presented for n = 10 and n = 20 Rydberg atoms at velocities $1 < v/v_e < 10$ where $v_e = n^{-1}$ a.u. Electric fields which ionize product Rydberg atoms in states $n' = n + \Delta n$ with $\Delta n = 1, 2$ and 4 were used. The electric field caused the cross sections for electron capture to decrease by up to fourfold while the ionization values increased by up to two orders-of-magnitude.

f. Submitted to J. Phys. B.

THE EFFECT OF CORE INTERACTIONS ON ℓ-MIXING COLLISIONS OF RYDBERG ATOMS WITH RARE GASES

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Coupled channel calculations for collisions of Na Rydberg atoms with He and Ar have been performed to investigate the effect of the interaction between the Na^+ core and the rare gas. For $n = 10$, channels corresponding to the levels nd and nf are included, and calculations are reported both with and without terms arising from the core. It is found that the inelastic cross sections (10d → 10f) are insensitive to the core interactions, whereas the elastic cross sections (10d → 10d) may change significantly. This result is consistent with the prediction of the first order Born approximation.

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